

Appendix A

Neptunium-237 Target Fabrication and Processing Operations for Plutonium-238 Production

This appendix includes a description of the Oak Ridge National Laboratory (ORNL) Radiochemical Engineering Development Center (REDC), the Idaho National Engineering and Environmental Laboratory (INEEL) Fluorine Dissolution Process Facility (FDPF), the Hanford Site (Hanford) Fuels and Materials Examination Facility (FMEF), and the proposed processing facilities and technologies that would be used to store neptunium-237, fabricate neptunium-237 targets, process irradiated targets for plutonium-238 production, recycle neptunium-237, and ship plutonium-238 oxide to Los Alamos National Laboratory (LANL). The material presented in this appendix is based primarily on *Preconceptual Design Planning for Chemical Processing to Support Pu-238 Production* (Wham et al. 1998), except where noted.

A.1 RADIOCHEMICAL ENGINEERING DEVELOPMENT CENTER

A.1.1 Facility Description

REDC is part of the Melton Valley 7900 Complex at ORNL. The High Flux Isotope Reactor (HFIR) is also part of the Melton Valley 7900 Complex. The REDC Complex includes a hot cell facility, Building 7930, which is specifically designed to address the problems associated with the containment of actinide element isotopes and their daughter isotopes. This facility was also designed to protect workers from high dose rates of penetrating radiation, including fast neutrons from spontaneous fission that require thick shielding and the capability for remote operation using manipulators. Building 7930, the proposed site for the plutonium-238 production project, has been used for a variety of transuranium actinide element projects, most notably the processing of californium-252, curium-244, and curium-248 for use as high-intensity neutron sources and research radioisotopes. The current californium-252 operations in Building 7930 would continue and coexist with the plutonium-238 production project.

Building 7930 is a three-story structure with a partial basement, constructed of structural steel, reinforced concrete, and masonry. Perimeter walls are reinforced concrete block. Floors are reinforced concrete slabs that are either poured on compacted aggregate or are supported by structural steel. The roof, replaced in the summer of 1997, is metal decking covered with built-up roofing. The building has a gross floor area of 3,062 square meters (32,950 square feet), exclusive of hot cells. The cell complex adds 286 square meters (3,080 square feet). The total enclosed volume is 18,295 cubic meters (646,000 cubic feet). The building is divided into four major areas: (1) the hot cell complex, consisting of six shielded cells and one unshielded cell; (2) maintenance and service areas; (3) an operating control area; and (4) an office area. Also included are utility services, ventilation systems, crane and manipulator systems, and liquid waste systems. The first and second floor plans of Building 7930 are shown in **Figures A-1** and **A-2**, respectively.

Cells D and E and space on both the second and third floors would be used for the plutonium-238 project. Cells D and E are both clean and empty and could be used for this work with minimal modifications. Cell D activities would include receipt of irradiated targets, target dissolution, chemical separation of neptunium and plutonium from fission products, partitioning and purification of the neptunium, and transuranic waste processing. Cell E would contain processing equipment to purify the plutonium-238, prepare plutonium oxide, and transfer the oxide into shipping containers. Cell E also would provide temporary storage of the neptunium oxide from the Savannah River Site (SRS). Cell F is also a possible interim location for storing neptunium. Neptunium-237 target fabrication would be completed on the second floor outside the cell, but inside a glovebox.

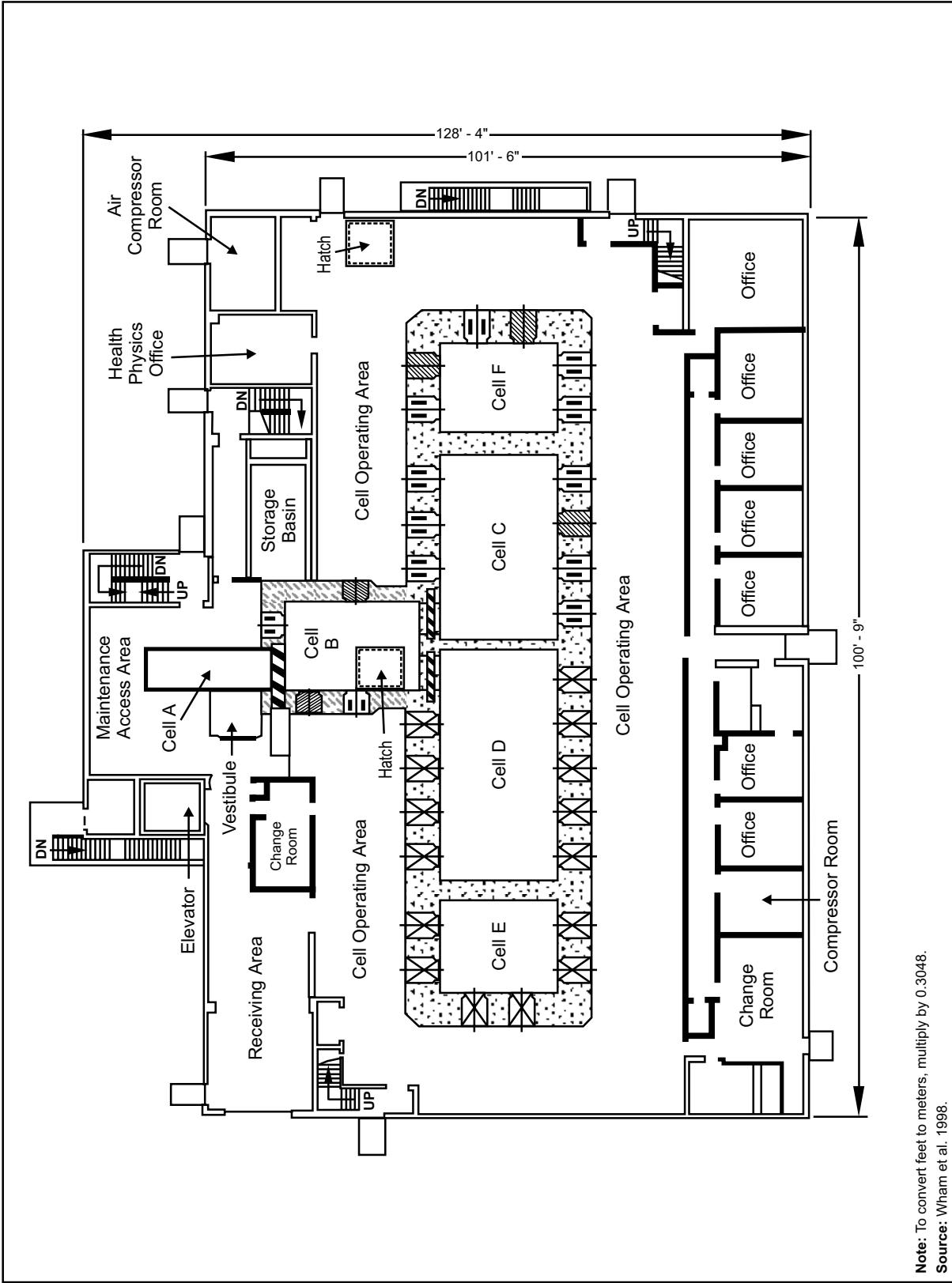


Figure A-1 First Floor Plan for Building 7930

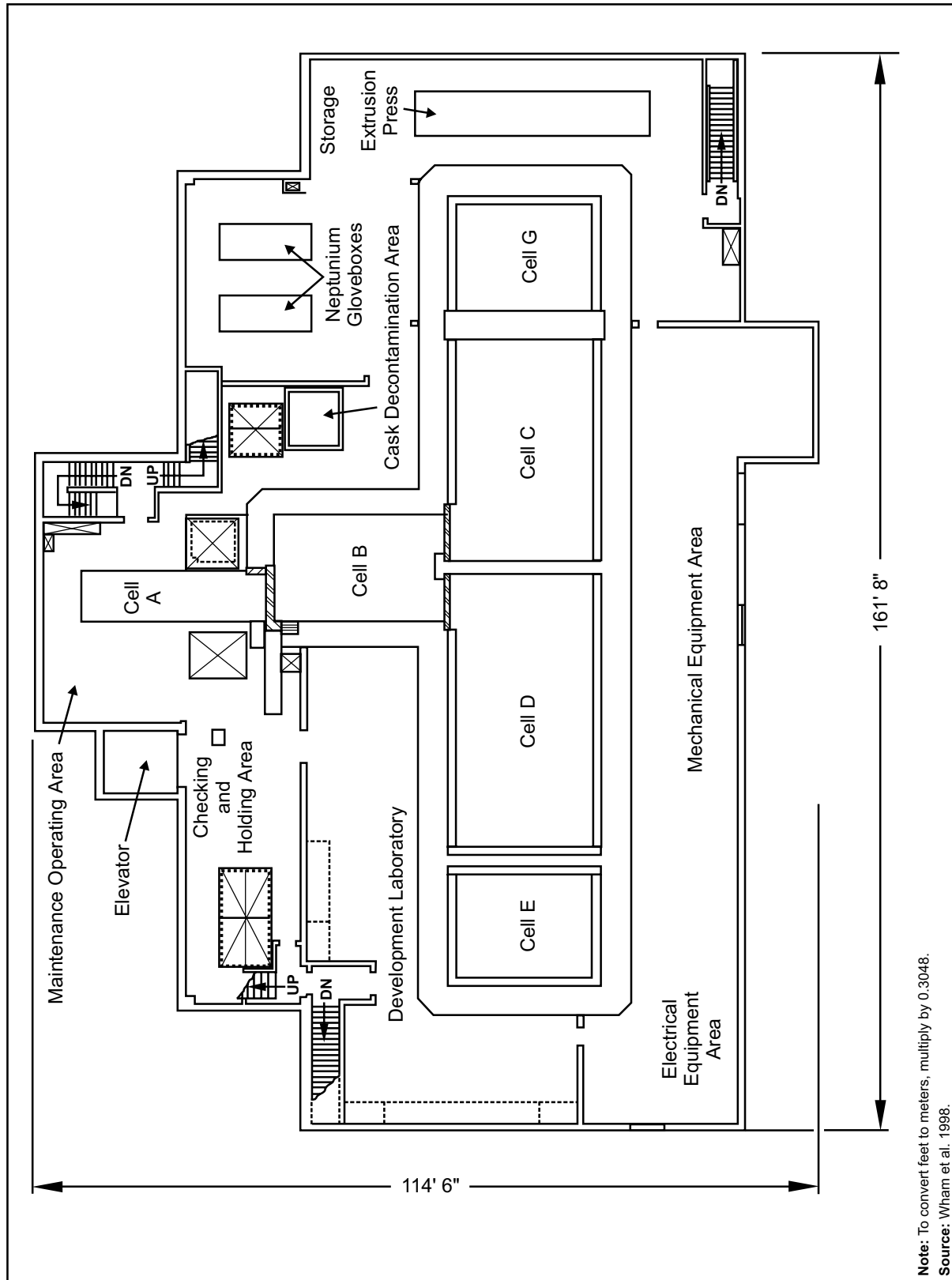


Figure A-2 Second Floor Plan for Building 7930

Cells D and E are adjacent and separated by a 1.2-meter-thick (4-foot-thick) shielding wall. The exterior walls are reinforced concrete 1.7 meters (5.5 feet) thick to a height of 3.4 meters (11 feet) above the first floor, and 1.4 meters (4.5 feet) thick from there to the roof, which is 1.5 meters (5 feet) thick. The cell floors are lined with Type 304L stainless steel, as are the walls and ceiling of Cell D. Currently, only the lower 0.3 meters (12 inches) of the Cell E walls are lined with stainless steel; above that, the walls are concrete with a 0.051-centimeter-thick (0.020-inch-thick) modified phenolic protective coating. For the plutonium-238 project, the walls in Cell E must be completely lined with stainless steel to improve containment and facilitate decontamination.

The lower section of each cell has operating modules, each with a large penetration for a viewing window and a pair of sleeves through which manipulators can be installed. Each cell is also equipped with transfer hatches and ports in the roof through which tools and equipment can be inserted.

Within a cell, operations would take place primarily with equipment contained in a workstation. A workstation is a stainless steel pan with short walls, and is open to the surrounding cell. The workstation has dimensions such that the manipulators can reach all equipment, and the entire area is viewable to the operators through the viewing window. A servomanipulator would be used to transfer materials between workstations. Transfer of items within workstations would be accomplished using the manipulators at each workstation.

Cell D has interior dimensions of 6.1 meters (20 feet) wide, 12.5 meters (41 feet) long, and 7.3 meters (24 feet) high. Currently, five chemical processing workstations and one analytical chemistry workstation are planned for Cell D. One window location would be used for transfer lines to bring in process solutions.

Cell E has interior dimensions of 6.1 meters (20 feet) wide, 4.9 meters (16 feet) long, and 9.1 meters (30 feet) high. A storage facility would be added, and three window locations would be used for chemical processing workstations. One of the workstations would be an enclosed stainless steel box. This workstation would be used for handling plutonium-238 oxide as a powder. Use of an enclosed area in this manner would minimize migration of plutonium-238 oxide powder and contamination.

All cells would be ventilated by air drawn from the occupied areas of the building through high-efficiency particulate air filters, and then through the cells on a once-through basis. The air leaving the cells would be filtered at the point of exit by high-capacity roughing filters, and then by two banks of high-efficiency particulate air filters in succession before being released to the atmosphere from Stack 7911.

Currently, Cells D and E have heat detectors, but are not equipped with fire suppression systems.

Target fabrication would be carried out on the second floor of Building 7930. The area is currently used as a maintenance shop and storage area. The walls between the maintenance shop and storage area would be removed to allow room for gloveboxes. The gloveboxes would be used to convert aqueous neptunium solutions into a form suitable for target fabrication. The storage area would be moved to another location within REDC and would be used for target fabrication equipment.

A.1.2 Neptunium-237 Storage

The neptunium-237 from SRS would arrive as neptunium dioxide, the most stable of the neptunium oxides. Upon arrival at REDC, the neptunium oxide would be removed from the shipping container(s). The product canister containing the neptunium oxide would remain in the containment vessel for storage in a designated shielded storage area until initiation of the purification process to remove protactinium-233.

After the neptunium-237 is processed into targets, the targets would be placed in shielded containers. The product canisters would be placed into containment vessels, and loaded directly into shipping containers for transport to the irradiation facility.

A.1.3 Neptunium-237 Target Fabrication Process Description

The fabrication of neptunium-237 targets for plutonium-238 production would require dissolving the neptunium oxide (if necessary), purification of neptunium to remove radioactive decay products, conversion of the neptunium to an oxide, and fabrication of neptunium oxide into targets for irradiation. The neptunium-237 to be used in the targets would come from two sources: (1) neptunium that had been separated previously during spent nuclear fuel processing at SRS, converted to an oxide, and then shipped to ORNL for storage (Section A.1.2); and (2) neptunium that would be recovered from irradiated neptunium-237 targets and recycled for use in new targets (Section A.1.4).

Initially, all of the neptunium-237 required for target fabrication would come from the neptunium oxide in storage. After postirradiation processing begins, most of the neptunium requirements would be met by using recycled neptunium, and only a small quantity of the stored neptunium would be needed to replace the neptunium transformed to plutonium-238 during irradiation.

All target fabrication activities at ORNL would be conducted in REDC Building 7930. The first stage of the target fabrication process would involve neptunium purification to remove protactinium-233 (a product of neptunium-237 alpha decay) and would be conducted in shielded facilities to minimize radiation exposure. Oxidation of the purified neptunium, mixing neptunium oxide with a suitable diluent, and preparing a billet for extrusion for Advanced Test Reactor (ATR) or HFIR targets, or preparing wafers or pellets for Fast Flux Test Facility (FFTF) targets, would be conducted in shielded gloveboxes on the second floor of Building 7930. The mechanical operations involved in the final target fabrication would be conducted in open boxes located in the target fabrication room.

The fabrication process for neptunium-237 targets is shown in **Figure A-3**. The neptunium would be brought from storage or recycling in preparation for purification. The neptunium would be dissolved (if necessary), treated to remove protactinium-233, and converted to an oxide. The oxide then would be transferred to the target fabrication line, where it would be formed into neptunium-237 targets.

A.1.3.1 Neptunium-237 Purification

Neptunium-237 is a radioactive isotope that decays to protactinium-233 through loss of an alpha particle. The protactinium reaches 90 percent of the equilibrium activity in approximately 10 weeks. Protactinium-233 has a short half-life (27 days) and decays to uranium-233, releasing gamma rays. The decay of this protactinium ingrowth would contribute significantly to the radiation doses to workers in the target fabrication

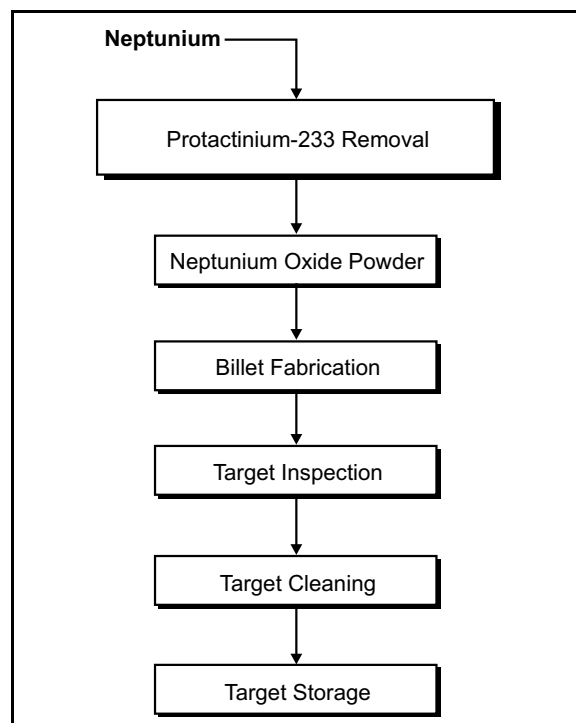


Figure A-3 Steps Required for Target Fabrication

line. Removal of the protactinium prior to oxide conversion and target fabrication would result in less radiation exposure for personnel involved in the target fabrication activities.

Neptunium oxide would be removed from its storage location in Cell E and transferred to dissolution equipment, which also would be located in Cell E. This dissolution may also take place in Cell D. The equipment would be sized to dissolve kilogram-size batches of neptunium-237. The neptunium would be dissolved in nitric acid and the solution would be passed through a column of silica gel, which would adsorb the protactinium. The purified neptunium-bearing solution, obtained in the third solvent extraction step during processing of irradiated neptunium-237 targets, would become the primary source of neptunium-237 for targets, once irradiated target processing began (Section A.1.4). Additional neptunium needed to supplement this source would come from the neptunium oxide stored in Cell E. The protactinium adsorbed in the silica gel then would be converted to a solid waste form. Because of the relatively short half-life of the protactinium-233, and its decay to uranium-233 (a relatively stable isotope), the radioactivity from this waste would be small after about 1 year of storage. The purified neptunium-237 solution would then be transferred to the target fabrication glovebox line for conversion of the neptunium to oxide.

A.1.3.2 Neptunium Oxide Production

The desired form of neptunium oxide for target fabrication is currently assumed to be oxide microspheres. The neptunium-237 solution would be passed through a cation-exchange column containing a resin, such as Dowex 50W-X8, of selected particle-size range (typically 60 to 80 microns [0.000024 to 0.000031 inches]). The loaded resin would be washed with dilute acid and dried with an air stream pulled through the column via a vacuum.

To convert the resin to oxide, a multistep heating cycle with ramp and hold times using both air and 4 percent hydrogen/argon streams would be employed. The preferred method would use heated air to burn the resin and to form the neptunium oxide microspheres. A typical cycle would be as follows: initially heat the resin with air (7 to 10 millimeters [0.28 to 0.39 inches] per second superficial velocity) at 150 °C (300 °F) for 1 hour; ramp to 450 °C (840 °F) at 5 °C (9 °F) per minute and hold for 1 hour with air; ramp to 800 °C (1,470 °F) at 10 °C (18 °F) per minute and hold for 4 hours with air (most of the carbon should be removed in this step); and switch to 4 percent hydrogen/argon and continue at 800 °C (1,470 °F) for a final 4-hour period to complete conversion to the oxide. The oxide then would be cooled for handling, transferred to a crucible, and sintered at 1,200 °C (2,190 °F) in air for 10 hours to complete the oxidation. This material would be weighed and characterized (tap density, radiochemical analysis, and particle size) to determine the blends for fabrication into pellets.

An alternative procedure for producing neptunium oxide would be precipitation of neptunium oxalate, followed by filtration and calcination to form neptunium oxide. The neptunium-bearing solution would be mixed with a solution containing oxalic acid. After mixing for at least 30 minutes, the supernatant would be decanted and filtered into a holding tank. The neptunium oxalate would be drained into a filter boat. The tank would be washed with 0.1 molar (*M*) oxalic acid and drained through the filter. The oxalate would dry on the filter and then be transferred to a platinum-lined furnace can. The filtrate would be sampled and sent to liquid waste treatment. The furnace can containing the neptunium oxalate would be placed into a calciner and heated to approximately 400 °C (750 °F) for 1 hour to decompose the neptunium oxalate to neptunium oxide and carbon dioxide.

A.1.3.3 Neptunium-237 Target Fabrication

Fabrication of the targets would take place in dedicated gloveboxes in Building 7930. The target for the ATR and HFIR reactors consists of the neptunium oxide blended with an inert filler such as aluminum powder, pressed into a target core, and clad with aluminum. This type of target has been used historically in nearly all of the U.S. Department of Energy (DOE) water-cooled and -moderated production and research reactors used to produce isotopes, including plutonium-238. Three different fabrication techniques could be employed in making these types of targets for ATR and HFIR:

- Blending the neptunium oxide and aluminum powders, pressing the mixture into individual pellets, loading the pellets into aluminum target tubes, and seal welding and hydrostatic compression of the tubes
- Blending the neptunium oxide and aluminum powders, pressing the mixture into compacts, roll milling the compacts between aluminum-clad material, and seal welding the aluminum-clad neptunium dioxide and aluminum plates
- Blending the neptunium oxide and aluminum powders; pressing the mixture into billets; assembling the billets into welded, evacuated containers; and coextruding the neptunium oxide and aluminum mixture with the aluminum container to produce target tubes

Another fabrication technique that may be considered is mixing the neptunium oxide with a high-temperature diluent (other than aluminum), pressing the mixture into pellets or tubes, and sealing it into Zircaloy tubing. Targets with Zircaloy or stainless steel cladding would be used in targets for the commercial light water reactor (CLWR) or a high-energy accelerator due to higher operating temperatures. This fabrication technique would use similar gloveboxes for target fabrication and quality assurance tests of the targets. The proposed target for the FFTF reactor would consist of alternating wafers or pellets of neptunium oxide and yttrium hydride (moderator) sealed in stainless or ferritic alloy steel tubing to make target pins. These pins, up to 2.5 centimeters (1 inch) in diameter by 2.4 meters (8 feet) long, would subsequently be inserted into an inlet nozzle, duct, and handling socket assembly for handling and insertion in the FFTF reactor. Fabrication and assembly operations would be done in shielded gloveboxes to minimize personnel radiation exposure. The completed targets then would be stored in Building 7930 until shipment to a reactor for irradiation.

A.1.4 Postirradiation Target Processing Description

Postirradiation processing of neptunium-237 targets at ORNL would involve dissolution; separation of the actinides from the fission products; separation of neptunium from plutonium; a third solvent extraction process to purify the remaining neptunium; purification of plutonium; precipitation of plutonium oxalate; calcination of plutonium oxalate to plutonium oxide; and exchange of oxygen-17 and -18 by oxygen-16 in the plutonium oxide. The dissolution and purification processes would be conducted in Cell D of Building 7930, and the plutonium purification, plutonium oxide preparation, oxygen exchange reaction, and transfer of the plutonium oxide to shipping containers would be conducted in Cell E of Building 7930.

The postirradiation target processing steps are shown in **Figure A-4**. The irradiated targets would be cooled at the irradiation site for at least 120 days to allow time for decay of short-lived fission products.

A.1.4.1 Target Dissolution

Irradiated targets from ATR or HFIR would be brought to Building 7930 in a Type B shipping cask with sufficient shielding to meet U.S. Department of Transportation (DOT) requirements and transferred to Cell D,

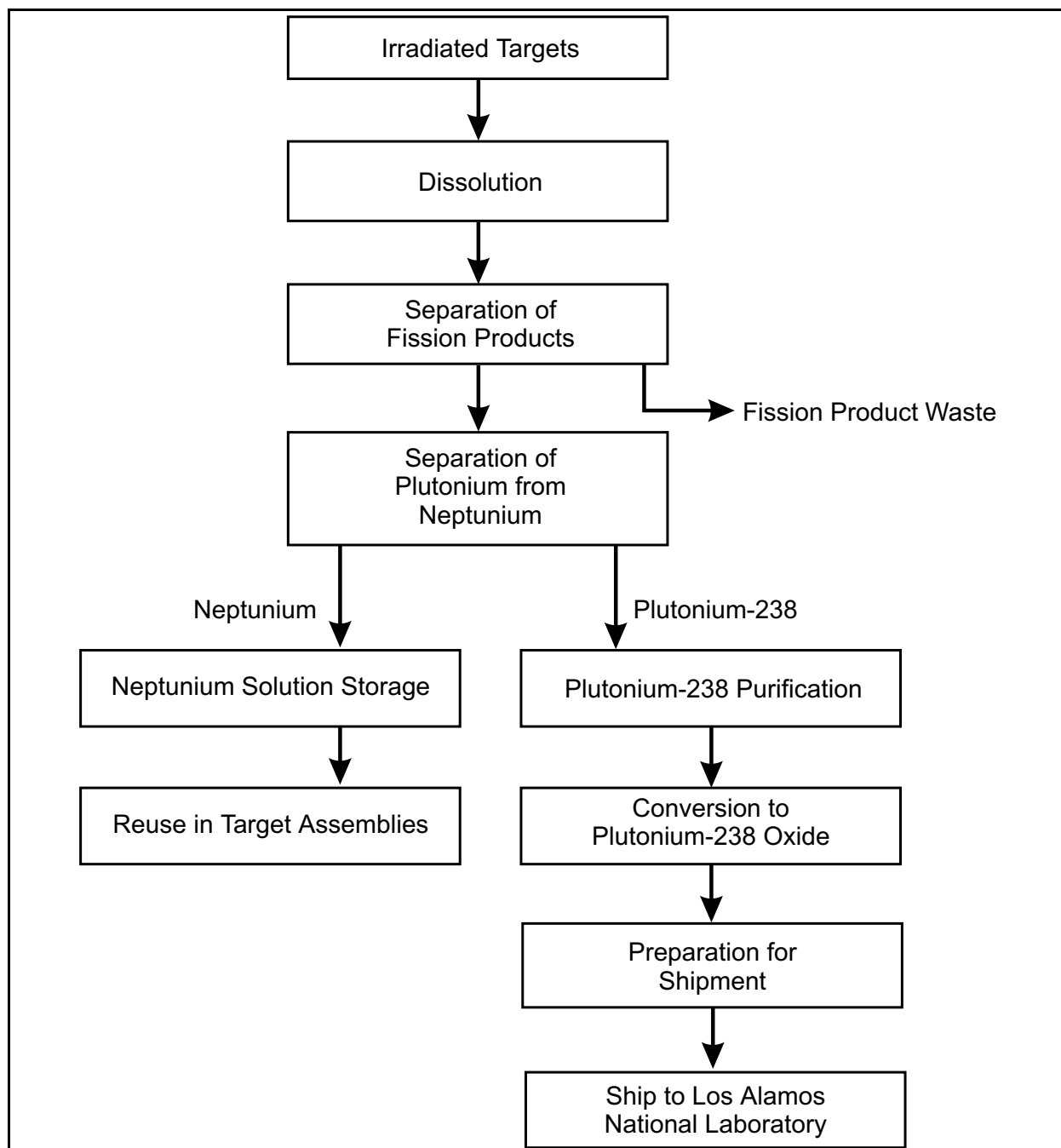


Figure A-4 Irradiated Neptunium-237 Target Processing

where postirradiation processing of the targets would be conducted. Dissolution of the aluminum-clad irradiated targets from HFIR or ATR would be accomplished using a two-step process. In the first step, the irradiated targets would be submerged in a 2.25 *M* sodium nitrate solution and heated to 90 to 95 °C (194 to 203 °F). A 10 *M* sodium hydroxide solution then would be added at a controlled rate to sustain the dissolution reaction. The aluminum-bearing caustic solution would be pumped through parallel sintered stainless steel filters and discarded as low-level waste. The filter then would be backflushed to the dissolver tank, where the remaining solids would be digested in a solution consisting of 8 *M* nitric acid and 0.02 *M* sodium fluoride. This solution would dissolve the actinides and most of the remaining fission products. This solution would

be filtered to remove any remaining solids (primarily silica fission products) and produce an acid product for solvent extraction.

Stainless steel- or Zircaloy-clad targets irradiated in the CLWR or in FFTF would be cut into small pieces and leached with nitric acid or other suitable solution to dissolve the neptunium, plutonium, and fission products away from the insoluble cladding. The solution would be filtered and the undissolved cladding would be discarded as low-level waste.

A.1.4.2 Plutonium Separation and Neptunium Recycling

The solution containing neptunium and plutonium would be processed in a series of three solvent extraction steps. In the first step, the neptunium and plutonium would be separated from fission products by extraction into an organic phase consisting of tributyl phosphate dissolved in normal paraffin hydrocarbon. Fission products and other contaminants would remain in the aqueous phase. After separation of the aqueous and organic phases, neptunium and plutonium would be stripped from the organic phase into another aqueous phase using a solution of 0.1 *M* nitric acid containing 0.1 *M* hydroxylamine nitrate as a reducing agent. Reduction would convert neptunium into the +4 oxidation state and plutonium into the +3 oxidation state. This solution then would flow to the second solvent extraction stage.

In the second solvent extraction step, neptunium (which would be in the +4 oxidation state) would be selectively extracted back into an organic phase consisting of 30 percent tributyl phosphate in normal paraffin hydrocarbon, while plutonium (which would be in the +3 oxidation state) would remain in the aqueous phase. Control of the oxidation states would be accomplished by the presence of hydroxylamine nitrate in the feed solution and the use of an aqueous hydroxylamine nitrate scrub stream to maintain the proper oxidation states within the solvent extraction contactor. Neptunium then would be stripped from the organic phase using a solution of 0.1 *M* hydroxylamine nitrate and 0.2 *M* nitric acid. The acidity of the neptunium-bearing solution would be adjusted to 2 *M* nitric acid and would be routed to a third solvent extraction step, where the solution would be further purified. The plutonium-bearing stream would be transferred to Cell E, where it would be further purified, if necessary, and converted to an oxide.

In the third solvent extraction step, any plutonium remaining in the neptunium-bearing solution would be separated from the neptunium. The neptunium-bearing solution from the second solvent extraction step would be mixed with a solution of 30 percent tributyl phosphate in normal paraffin hydrocarbon and a solution of 0.1 *M* nitric acid and 0.1 *M* ferrous sulfamate. The ferrous sulfamate would act as a reductant to ensure that the plutonium would remain in the +3 oxidation state and in the aqueous phase. The neptunium would be extracted into the organic phase. After separation of the aqueous and organic phases, neptunium would be stripped from the organic phase using a solution of 0.2 *M* nitric acid and 0.1 *M* hydroxylamine nitrate. The purified neptunium-bearing solution then would be stored in a tank below Cell D until needed for conversion to an oxide for fabrication into targets, as described in Section A.1.3.

A.1.4.3 Plutonium Purification and Preparation of Plutonium Oxide

Although the necessity for further plutonium purification from the second solvent extraction has not yet been fully determined, provisions would be made to purify the plutonium-bearing solution using an anion exchange process. This process has been used previously at REDC to purify plutonium products in preparation for precipitation and calcination to an oxide product. If the plutonium product solution from the second-cycle solvent extraction process meets the desired specifications, no anion exchange processing would be needed and the solution would be sent directly to the oxalate precipitation process. If purification were required, the feed solution would be adjusted to a high acid concentration (approximately 8 *M* nitric acid) and the oxidation states of the actinides would be adjusted to form the metal nitrate complex that loads on the anion exchange

resin. The oxidation state of the metals would be adjusted using a reductant such as ferrous sulfate or ferrous sulfamate. After loading the plutonium onto the resin, the resin would be washed with an acid solution to remove impurities, and the plutonium would be selectively stripped with a dilute acid solution or a dilute acid solution containing reductants (hydroxylamine nitrate, nitrous acid, or hydrazine) to reduce the oxidation state of the loaded complex and strip it from the anion resin column.

The plutonium product from the second-cycle solvent extraction process or the anion exchange process would be adjusted to give a final solution of 1 *M* nitric acid with plutonium in the +4 oxidation state. Ascorbic acid would be used as the reductant to adjust the plutonium oxidation state because it would not add any extraneous ions to the final product upon conversion to the oxide. Two additions of 1 *M* oxalic acid would be made to the plutonium solution to quantitatively precipitate plutonium oxalate. The solution would be filtered and the plutonium oxalate collected on a sintered platinum/Inconel filter for calcination to the oxide. The precipitated plutonium oxalate and filter then would be transferred to a furnace and calcined in air at 735 °C (1,355 °F) for 2 hours to produce plutonium oxide.

The alpha-neutron reaction, which occurs when alpha particles emitted from plutonium-238 interact with atoms of naturally occurring oxygen, results in a high neutron emission rate from plutonium oxide, and may cause a high neutron exposure to workers. To reduce this exposure, an oxygen exchange process would be used to replace the higher cross-section oxygen isotopes (oxygen-17 and oxygen-18) with oxygen-16, which has a very small cross section for the alpha-neutron reaction. To accomplish this exchange, a stream of oxygen-16 enriched gas would be passed through the plutonium oxide product from the calcination step, above, at a temperature of approximately 800 °C (1,470 °F) for approximately 4 hours. The progress of the exchange reaction would be constantly monitored with a neutron detector located adjacent to the plutonium oxide. After completion of the exchange reaction, the plutonium oxide would be cooled in an inert atmosphere and immediately transferred to a container (e.g., EP-60) for final packaging (see Section J.3.3.3 for a further discussion of this packaging system).

A.1.5 Plutonium-238 Storage Description

A container (e.g., EP-60) containing plutonium-238 would be placed into a primary containment vessel (e.g., EP-61), which then would be placed into a secondary containment vessel (e.g., EP-62) and stored until shipment to LANL. The secondary containment vessel then would be loaded into a shipping package (e.g., 5320B). DOE anticipates about four shipments per year to LANL, as described in Appendix J.

A.2 FLUORINEL DISSOLUTION PROCESS FACILITY

A.2.1 Facility Description

The Idaho Nuclear Technology and Engineering Center (INTEC) is located northeast of the Central Facilities Area at INEEL and approximately 3.2 kilometers (2 miles) southeast of ATR. Two buildings at INTEC are proposed storage and processing sites for plutonium-238 production: Building CPP-651, the Unirradiated Fuel Storage Facility, and Building CPP-666, FDPF and Fuel Storage Facility.

Building CPP-651 was originally designed for the storage of special nuclear materials to support Defense Programs and is quite flexible in terms of the size and shape of special nuclear materials that it can receive and store. The 100 storage positions in the vault use the existing structural barriers of Building CPP-651 (earth and concrete) and provide supplemental security protection via their in-ground concrete storage silo design. Each storage position houses a rack that holds seven highly enriched uranium product cans. Racks are raised and lowered in their storage positions via an overhead 1-ton hoist.

Building CPP-666 is divided into two parts, the Fuel Storage Facility and FDPF. The Fuel Storage Facility consists of receiving and unloading areas, a fuel unloading pool, and six storage pools for storing nuclear fuel.

FDPF was designed and built to process Navy fuel via three dissolver trains. When fuel reprocessing was discontinued, uranium and hazardous materials were flushed from FDPF, and the facility is currently under consideration for new missions. FDPF consists of a large hot cell and supporting areas with a total area of approximately 3,700 square meters (40,000 square feet). The facility is divided into five levels identified by their elevation relative to ground level (Hochhalter 1982). A floor plan of the +28-foot level, the proposed location for the neptunium glovebox train and the target fabrication glovebox, is shown in **Figure A-5**.

The FDPF cell is approximately 6 meters (20 feet) wide, 30 meters (100 feet) long, and 15 meters (50 feet) deep (Sire et al. 1992) with 1.8-meter-thick (6-foot-thick) concrete walls. A plan view of the FDPF cell is shown in **Figure A-6**. The cell includes manipulators, three dissolvers, off-gas cleanup systems, complexing vessels, process makeup vessels, pumps, valves, piping, and instrumentation (Hochhalter 1982). The interim storage rack area located in the south end of the cell could be used to store irradiated targets. A new 12.5-liter (3.3-gallon) or 200-liter (53-gallon) dissolution vessel would need to be installed. This dissolution vessel would be located in proximity to the existing Train One dissolver to facilitate the use of the existing dissolver off-gas system (Kirkham 1999).

A.2.2 Neptunium Storage

Neptunium-237 oxide would be shipped from SRS to INEEL in double-sealed containers loaded in Type B packages. At INEEL, it would be stored in the Building CPP-651 vault. There are 100 in-ground concrete-shielded storage positions (each approximately 25 centimeters [10 inches] in diameter by 2.4 meters [8 feet] in length) in this vault. Each storage position houses a rack that holds seven highly enriched uranium product cans, with each can containing about 8 to 10 kilograms (18 to 22 pounds) of highly enriched uranium (the total mass of the rack and cans is about 148 kilograms [325 pounds]). The design-basis radiation level for these cans is about 800 millirem per hour at contact. The rack that fits into the storage position can be redesigned for neptunium oxide containers if its existing dimensions are not adequate. Alternatively, neptunium-237 oxide may be stored in the processing hot cell (Cook and Hill 1999).

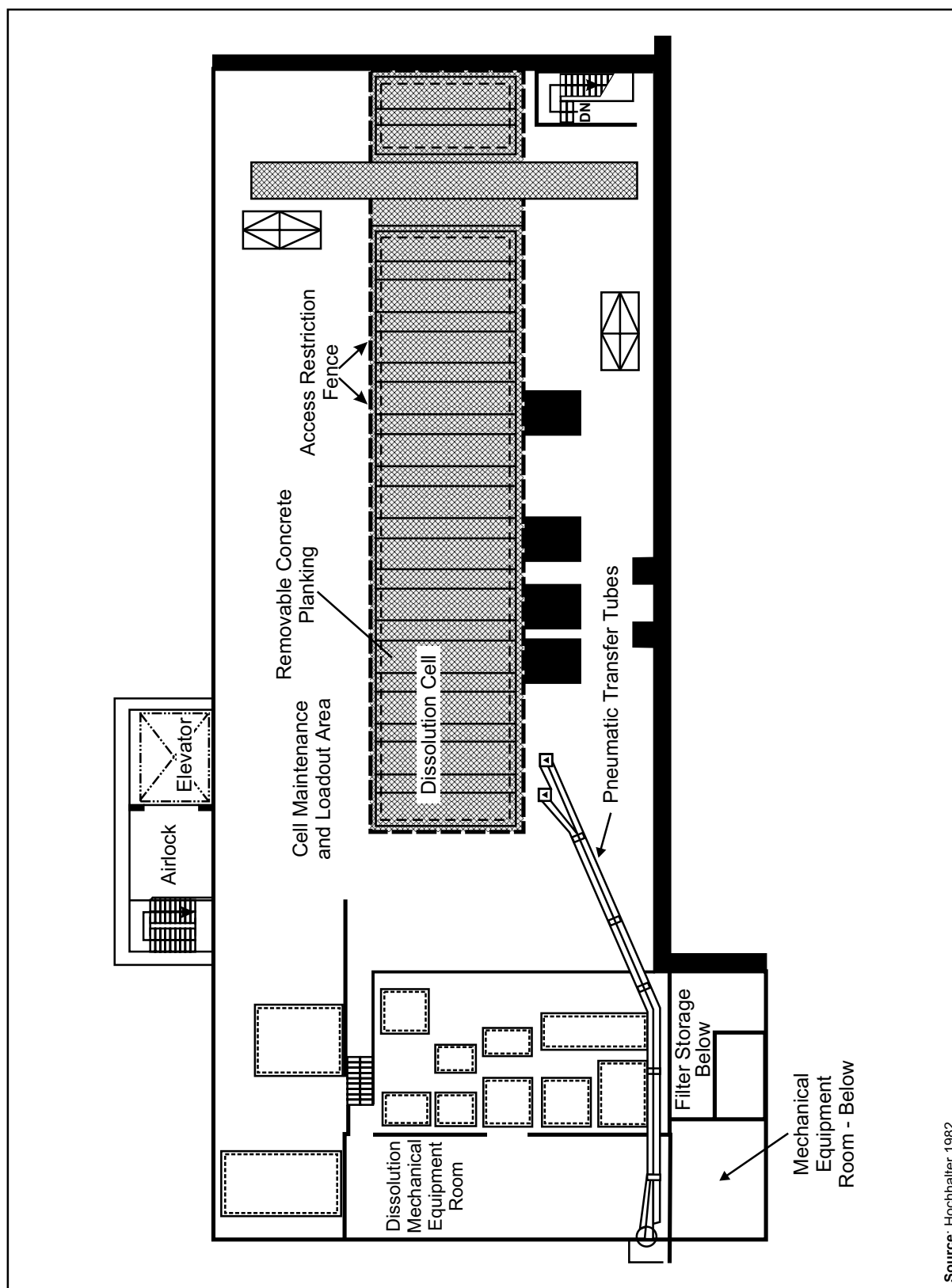
A.2.3 Neptunium-237 Target Fabrication Process Description

The neptunium-237 target fabrication process used at INEEL would be similar to that described in Section A.1.3 for target fabrication in REDC at ORNL and in more detail in *Preconceptual Design Planning for Chemical Processing to Support Pu-238 Production* (Wham et al. 1998). In addition to the target design considered in that document, INEEL could produce targets suitable for a commercial reactor or for an accelerator. The latter target is significantly longer than those considered by ORNL. The target fabrication process would include neptunium-237 purification, neptunium oxide production, and target fabrication.

A.2.3.1 Neptunium-237 Purification

Neptunium-237 oxide retrieved from storage would be purified by dissolving the material in nitric acid and passing the neptunium nitrate solution through a silica gel bed. During this process, the protactinium would adsorb onto the silica gel, leaving a protactinium-233-free neptunium nitrate solution. This process would either be conducted in the hot cell near the third-cycle solvent extraction equipment or within the neptunium processing glovebox train at the +28-foot level.

Neptunium nitrate recovered from postirradiation processing should not be contaminated by protactinium-233, as this isotope should have been removed in the solvent extraction process. However, if neptunium-237 target



Source: Hochhalter 1982.

Figure A-5 Fluorinel Dissolution Process Facility +28-Foot Level

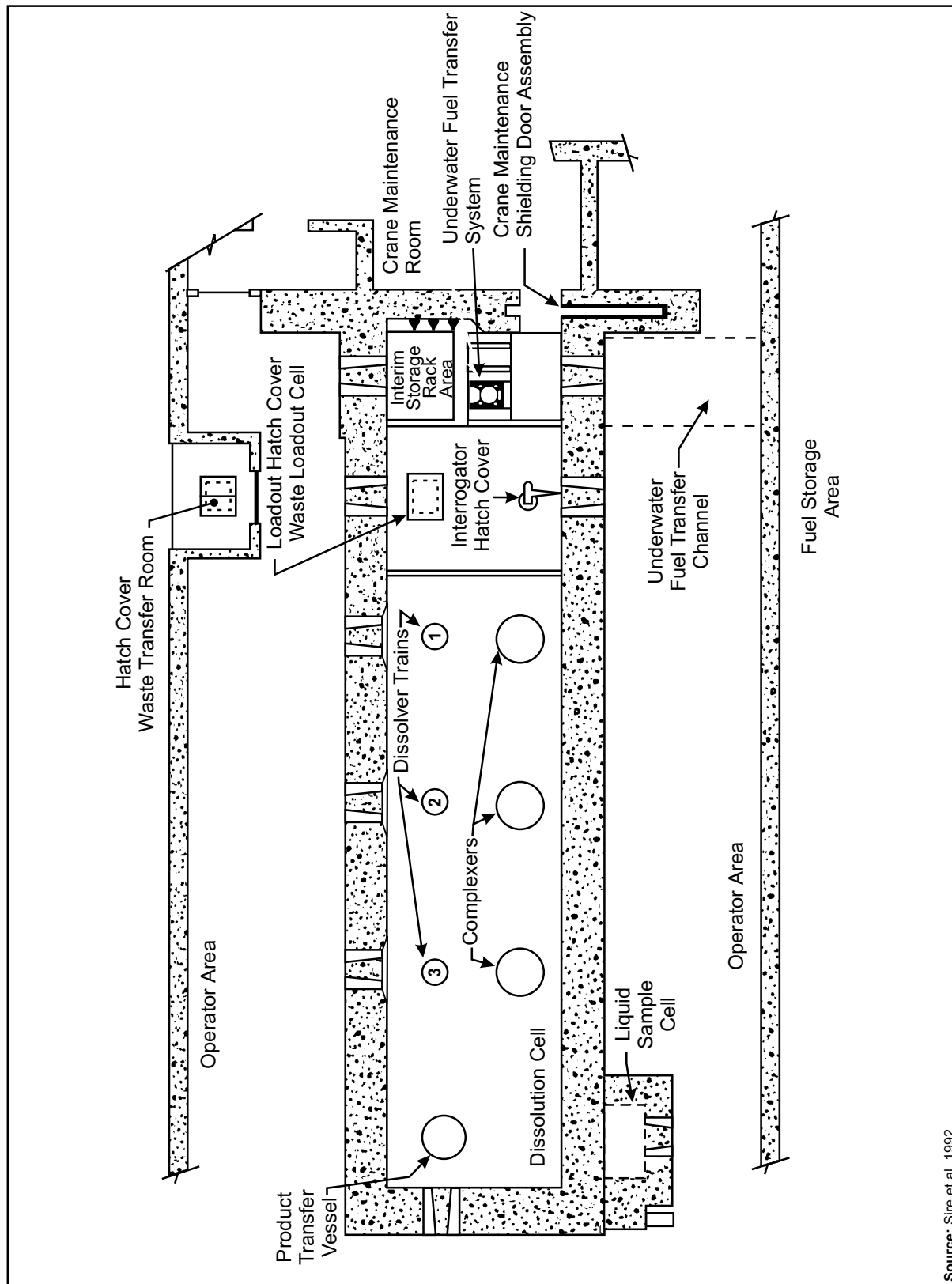


Figure A-6 Plan View of the Fluorinel Dissolution Process Facility Cell

Source: Sire et al. 1992.

fabrication were delayed, the ingrowth of protactinium-233 would require that the solution be passed through a silica gel bed to remove the protactinium prior to formation of neptunium oxide (Kirkham 1999).

A.2.3.2 Neptunium Oxide Production

Neptunium oxide production would be accomplished using the same process described for ORNL (Section A.1.3.2 and Wham et al. 1998). Conversion of the purified neptunium nitrate solution would be accomplished by adsorption of the neptunium on a cation resin bed in a glovebox train located on the cell maintenance level (+28-foot level). Solutions from the hot cell would be pumped from in-cell storage through a K-plug installed in an existing penetration. The routing would be through doubly contained lines run through the +17- and +28-foot levels from the operating level (0-foot level). (Note: The glovebox train may be located on any level on either side of the cell. The +28-foot level has ample area available for the glovebox train; however, it may be desirable to locate the glovebox train elsewhere because of better shielding and protection to the outside environment.) The resin then would be calcined to create microspheres of neptunium dioxide powder of the required morphology. All waste solutions from this process would be transferred to the cell waste collection system. The off-gas from the glovebox train would be filtered with a high-efficiency particulate air filtering system and tied into the existing cell off-gas system (Kirkham 1999).

A.2.3.3 Neptunium-237 Target Fabrication

Neptunium-237 target fabrication would be accomplished using the same target fabrication and assembly methods described for ORNL (Section A.1.3.3 and Wham et al. 1998). The proposed target design for ATR or HFIR consists of neptunium dioxide blended with aluminum powder, pressed into a target core, and clad with aluminum. The target used for ATR would be similar in appearance, but longer than the target that would be used for HFIR. (The ATR target length would be sized for the 1.2-meter [4-foot] active core length of ATR rather than the 51-centimeter [20-inch] active length of HFIR.) Target fabrication would be performed in an area adjacent to the neptunium glovebox train on the +28-foot level. The target fabrication operations would be performed in a shielded glovebox to provide adequate personnel protection from the varying dose rate. The target pins would be transferred to a shielded glovebox where they would be assembled into a target assembly. When completed and accepted, a target assembly would be removed from the glovebox, undergo required nondestructive assay analysis and verification, and be moved by elevator to the ground level and then by vehicle to the use location or to storage in the security area (CPP-651) (Kirkham 1999).

A.2.4 Postirradiation Processing Description

Postirradiation processing of neptunium-237 targets would use a similar process to the one described for ORNL in Section A.1.4 and in more detail in the preconceptual design study (Wham et al. 1998). An exception to this process is that aluminum-clad target dissolution would use a one-step dissolution using a nitric acid-fluoroboric acid solution instead of the two-step process that would be used by ORNL. There is adequate shielded cell floor space in FDPF to do the recovery operation.

A.2.4.1 Target Dissolution

Irradiated targets would be unloaded in the Fuel Storage Facility pool from the transfer cask. (This could be one of several approved existing casks used to transfer spent nuclear fuel.) The target container would be loaded under water into the transfer cart and moved up the incline channel into the FDPF cell. The targets may be placed in interim storage in the existing fuel storage ports in the cell or loaded directly into the dissolver. The dissolution would take place in a new 12.5-liter (3.3-gallon) vessel with an overflow into an existing dissolver vessel for continuous dissolution. Alternatively, a new vessel with about a 200-liter (53-gallon) capacity could be used for a batch process. This equipment would be located near the existing Train One

dissolver. Each aluminum-clad target from ATR or HFIR would be dissolved in 120 liters (31.7 gallons) of 7.5 *M* nitric acid and 0.15 *M* fluoroboric acid at 100 °C (212 °F). This dissolution results in the neptunium and plutonium being highly complexed with fluoride. Therefore, to perform a solvent extraction separation, the fluoride would have to be complexed to free the actinide ions. This could be accomplished by adding zirconium (IV) nitrate (Cook and Hill 1999).

At ORNL and Hanford, targets with a stainless steel or Zircaloy cladding (e.g., CLWR, FFTF, or high-energy accelerator targets) would be cut into pieces and leached with nitric acid or another suitable solution to dissolve the neptunium, plutonium, and fission products away from the insoluble cladding. The solution would be filtered and the undissolved cladding would be discarded as waste. At INEEL, the stainless steel- or Zircaloy-clad targets would be dissolved in a one-step process using a suitable solution.

A.2.4.2 Plutonium Separation and Neptunium Recycling

The process solution would be transferred by a positive displacement pump to the solvent extraction system, which would use centrifugal contactors. The solvent extraction system would be located at the south end of the cell near manipulators. Crucial components would be skid-mounted and within reach of manipulators. Because of the small flow rates needed, reagents would be fed into the separation system from small feed vessels located on the +28-foot level or in the operating corridor. Chemical makeup would take place in the existing makeup area with transfers to these smaller vessels. The acidic target solution would be treated with tributyl phosphate dissolved in normal paraffin hydrocarbon in three trains of centrifugal contactors and the plutonium and neptunium would be extracted into the tributyl phosphate/normal paraffin hydrocarbon phase.

Four separate transfer lines connect the FDPF hot cell with Building CPP-601, where the waste can be transferred to Building CPP-604. Building CPP-604 houses the Process Waste Evaporator, which would be used to evaporate the liquid waste for subsequent disposal. Aqueous waste streams would be collected in one of the existing complexer vessels, transferred to the existing product transfer vessel, and bled off to the Process Equipment Waste Evaporator system. An intercycle evaporator might be necessary to concentrate the first cycle strip before the second cycle. It would be located in the cell area south of and near the Train One dissolver or complexer vessel and suspended below the grating at the 0-foot level. The condensate would be routed to waste collection, with the concentrated solution going to the second solvent extraction cycle to separate neptunium and plutonium from each other. This extraction cycle also would use centrifugal contactors. The partitioned neptunium would go to the third solvent extraction cycle where it would be purified of any remaining fission products and stored for conversion to neptunium oxide (Section A.2.3). The plutonium-bearing solution would be purified by ion exchange and stored in the cell for subsequent processing to plutonium oxide.

A.2.4.3 Preparation of Plutonium Oxide

The conversion of plutonium nitrate solution to plutonium oxide would be accomplished in the same manner identified in the ORNL preconceptual design study for plutonium-238 production support operations (Wham et al. 1998). Plutonium oxide conversion would occur in shielded gloveboxes on the +28-foot level. The plutonium nitrate solution would be treated to adjust the plutonium oxidation state and then precipitated as an oxalate. The plutonium oxalate would be washed with dilute acid and calcined at the required temperature. The required oxygen-16 exchange would be done as part of the calcination step. The oxide would be packaged in a manner to assure maintenance of the desired degree of oxygen-16 exchange. The packaged material would undergo nondestructive assay analysis for accountability control and placed into storage pending packaging for shipment.

A.2.5 Plutonium-238 Storage Description

Plutonium-238 oxide would be transferred to the security area in Building CPP-651 for storage prior to shipment to LANL. Storage quantities would be expected to be small, as shipments would be made on a regular basis.

A.3 FUELS AND MATERIALS EXAMINATION FACILITY

A.3.1 Facility Description

FMEF is located in the 400 Area on Hanford adjacent to the FFTF. Constructed in the late 1970s and early 1980s to perform fuel fabrication and development, and postirradiation examination of breeder reactor fuels, FMEF is being maintained in a condition suitable for a future mission. The building is clean and uncontaminated, as no nuclear materials have been introduced. FMEF has been well maintained for potential future missions (Hoyt et al. 1999).

FMEF consists of the Process Building with an attached Mechanical Equipment Wing on the west side and an Entry Wing across the south side of the building. The Mechanical Equipment Wing houses facility utility and support equipment. The Entry Wing provides space for reactor fuel assembly, a lunchroom, change rooms, a security station, office space, and administrative support areas (DOE 1995).

The Process Building is 53.3 meters (175 feet) wide by 82.3 meters (270 feet) long and extends from 10.7 meters (35 feet) below grade to 29.7 meters (98 feet) above grade. The total operating space is approximately 17,400 square meters (188,000 square feet). The building is divided into six operating floors, or levels, which are identified by their elevation relative to ground level (DOE 1995). The Process Building contains several large interconnected hot cells and many smaller connected hot cells. However, most cranes, windows, and manipulators were not installed because construction was halted prior to completing work on the hot cell complex (Hoyt et al. 1999).

FMEF has the physical attributes required to process, handle, and store large quantities of special nuclear material. It is a massive, reinforced-concrete, hardened structure with safety-related equipment and systems, designed as a seismic Category 1 to withstand the Hanford design-basis earthquake, tornado, high-wind, and volcanic ashfall events. FMEF was also designed to meet the physical safeguards and security requirements for processing and storing Category 1 quantities of special nuclear material (Hoyt et al. 1999).

Ample space exists in FMEF for plutonium-238 production support, and numerous facility configurations are possible. In the absence of a detailed engineering study, it was decided that the process support would be located at the -35-foot level using the process support cells to house the irradiated target processing. This configuration also would contain this project, with its relatively modest requirements, to as few levels as possible. Alternative facility configurations can be found in the *Summary of Strategy for Implementing Plutonium-238 Production Support Activities in FMEF* (Hoyt et al. 1999). A floor plan of the -35-foot level is shown in **Figure A-7**.

The shipping and receiving bay located on the 0-foot level would be used to support the shipment and receipt of safe, secure trailer/SafeGuards Transports and irradiated target cask transporters. Additional facilities on the 0-foot level would be used to transfer irradiated targets into the storage area, decontaminate, and prepare equipment for maintenance, and package remote-handled solid waste for disposal. On the -17-foot level, the entry tunnel transporter would be used, as well as existing facility systems, as needed (Hoyt et al. 1999).

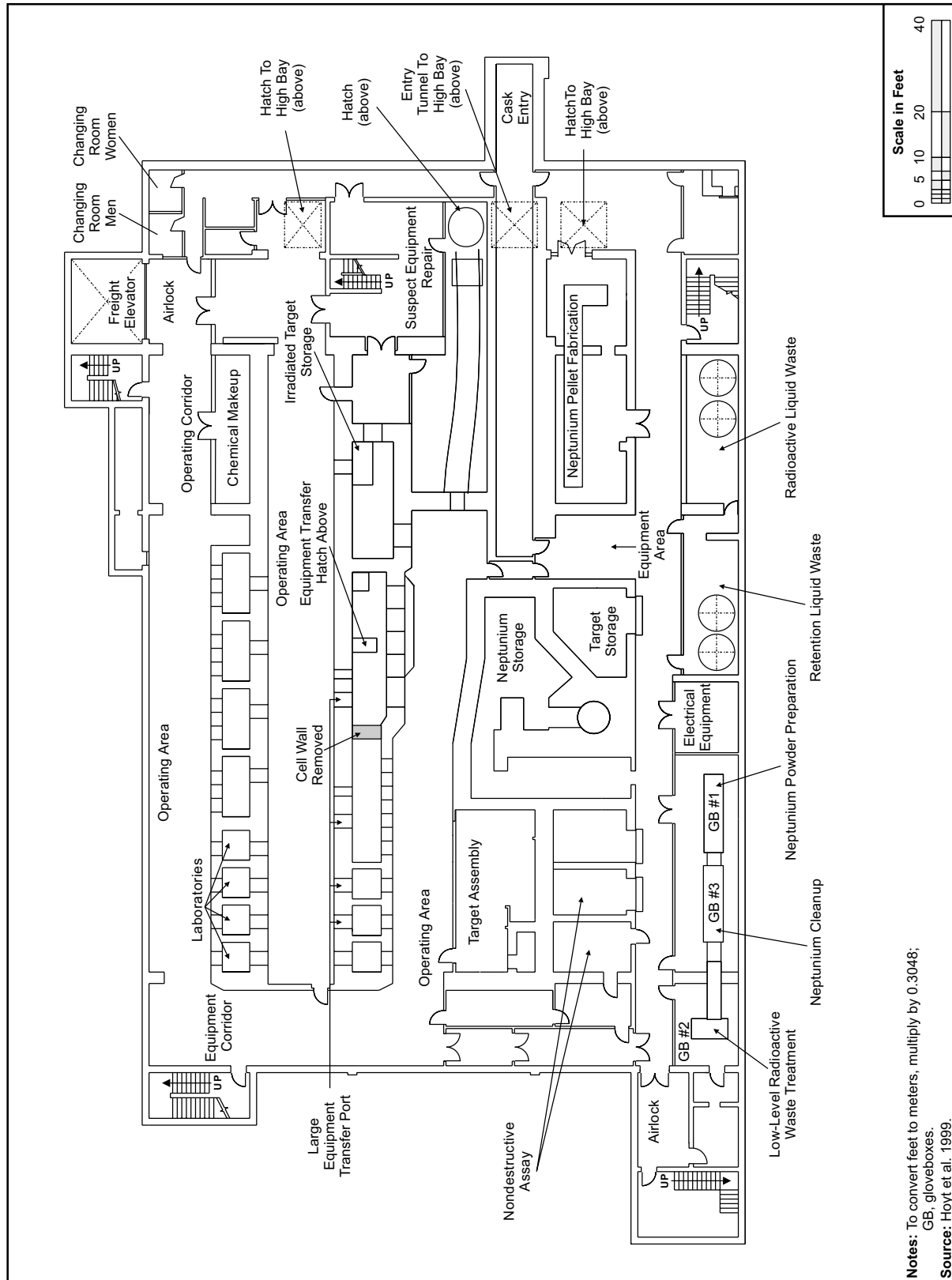


Figure A-7 Layout of the Fuels and Materials Examination Facility

The -35-foot level would house most of the processing and storage functions for plutonium-238 production. Neptunium-237 storage, target fabrication, and assembly would be located in rooms on the south side of the -35-foot level and include Rooms 112, 114, 124, and 128 (Hoyt et al. 1999).

The south bank of process support cells would be dedicated to target processing. Located on the -35-foot level, the 14 process support cells are arranged in two parallel rows along a horizontal transfer corridor. The process support cell complex is approximately 12.1 meters (40 feet) wide by 30.2 meters (99 feet) long. With the exception of Cell 146, each of the process support cells is 4.3 meters (14 feet) high and lined with stainless steel. Cell 146 extends to the 0-foot level and would be lined with stainless steel for the proposed project. The process support cell area is heavily shielded with either 122 centimeters (48 inches) or 81 centimeters (32 inches) of high-density concrete. Work in the cells would be performed using remotely operated equipment (DOE 1997).

Irradiated neptunium targets would be lowered through a hatch into Cell 147 and stored awaiting processing. Target processing would begin in Cell 146 and proceed through to plutonium-238 oxide conversion, storage, and loadout in Cell 142. The main target processing activities would occur in Cell 146. Existing wastewater collection systems would be used, and hot repair facilities also would be available on this level (Hoyt et al. 1999).

A.3.2 Neptunium Storage

Neptunium-237 oxide would be shipped from SRS to Hanford in double-sealed containers loaded in Type B packages. At Hanford, it would be stored in a vertical tube rack located in Room 114 on the -35-foot level of FMEF. This room was designed earlier to contain the TRIGA (training, research, isotopes General Atomics) reactor spent nuclear fuel and would provide excellent shielding capabilities. The room would be modified to provide storage racks to hold the 3013 containers. Individual 3013 container transfers would be accomplished remotely with an overhead crane to minimize personnel exposure.

A.3.3 Neptunium-237 Target Fabrication Process Description

The neptunium-237 target fabrication process used at Hanford would be similar to that described in Section A.1.3 for target fabrication in REDC at ORNL and in more detail in *Preconceptual Design Planning for Chemical Processing to Support Pu-238 Production* (Wham et al. 1998). In addition to the target designs considered in that document, Hanford could produce targets suitable for FFTF, a commercial reactor, or an accelerator. The latter target is significantly longer than those considered by ORNL. The target fabrication process would include neptunium-237 purification, neptunium oxide production, and target fabrication.

A.3.3.1 Neptunium-237 Purification

Neptunium-237 oxide retrieved from storage would be purified by dissolving the material in nitric acid and passing the neptunium-nitrate solution through a silica gel bed. During this process, the protactinium would adsorb onto the silica gel, leaving a neptunium nitrate solution that is free of protactinium-233.

For neptunium nitrate recovered from postirradiation processing, the protactinium-233 should have been removed in the solvent extraction process. However, if any additional purification were required, this would be achieved either through an anion exchange or an oxalate precipitation process. If oxalate precipitation were to be used, the neptunium oxalate precipitate would be washed to remove entrained liquor, and redissolved in nitric acid (Kirkham 1999).

A.3.3.2 Neptunium Oxide Production

Conversion of the purified neptunium nitrate solution would be accomplished by adsorption of the neptunium on a cation resin bed. The resin then would be calcined to create microspheres of neptunium dioxide powder of the required morphology.

A.3.3.3 Neptunium-237 Target Fabrication

Neptunium-237 target fabrication would be accomplished using the same target fabrication and assembly methods described for ORNL (Section A.1.3.3 and Wham et al. 1998). Target fabrication would be performed in FMEF Room 124, which would be modified by removing a wall in the area to create sufficient unencumbered floor space. The target fabrication operations would be performed in a shielded glovebox to provide adequate personnel protection from the varying dose rate. The target pins would be transferred to a shielded glovebox in Room 128, where they would be assembled into a target assembly. When completed and accepted, a target assembly would be removed from the glovebox, undergo required nondestructive assay analysis and verification, and be packaged for interim storage in Room 112 on the -35-foot level pending shipment to the reactor site. Fuel storage tubes located in the Fuel Assembly Area also could be used. Final selection would depend on protection requirements and the optimized building configuration (Kirkham 1999).

A.3.4 Postirradiation Processing Description

Postirradiation processing of neptunium-237 targets would use the same process described for ORNL in Section A.1.4 and in more detail in the preconceptual design study (Wham et al. 1998). There is adequate shielded cell floor space on the -35-foot level in FMEF to do the recovery operation. The cells would have either 122 centimeters (48 inches) or 81 centimeters (32 inches) of high-density concrete as shielding walls.

A.3.4.1 Target Dissolution

The targets would be transferred from their storage position in Cell 147 to the dissolving cell, Cell 146. There the extraneous hardware would be removed and the target pins would be sheared into small segments and placed into a dissolver vessel located near the shear in Cell 146. Segments from targets irradiated in HFIR or ATR would be agitated in a caustic solution to dissolve the aluminum cladding and aluminum target diluent. After removal of the caustic aluminum waste solution, the neptunium target would be treated with nitric acid to dissolve the neptunium, plutonium, and fission products.

Stainless steel- or Zircaloy-clad targets irradiated in the CLWR, FFTF, or the high-energy accelerator would be cut into small pieces and leached with nitric acid or other suitable solution to dissolve the neptunium, plutonium, and fission products away from the insoluble cladding. The solution would be filtered and the undissolved cladding would be discarded as waste.

A.3.4.2 Plutonium Separation and Neptunium Recycling

The acidic target solution would be treated with tributyl phosphate dissolved in normal paraffin hydrocarbon in a mixer-settler vessel and the plutonium and neptunium would be extracted into the tributyl phosphate/normal paraffin hydrocarbon phase. The aqueous phase containing the mixed fission products would be treated again to remove trace quantities of neptunium and plutonium. Waste-handling equipment would be used to minimize the activity in low-level radioactive liquid waste and to stabilize solid waste into an acceptable waste form. This equipment would be included in the hot cells used for the chemical processing of irradiated targets for plutonium-238 production. The neptunium and plutonium would be extracted from the tributyl phosphate/normal paraffin hydrocarbon solution as an aqueous nitrate solution. The neptunium

and plutonium next would be separated from each other by additional solvent extraction and stored in their respective storage tanks pending further purification and conversion to oxides.

Alternatively, an anion exchange process may be added to the existing FMEF for separating fission products and purifying neptunium-237 and plutonium-238 from irradiated targets using a series of ion exchange columns.

A.3.4.3 Preparation of Plutonium Oxide

The conversion of plutonium nitrate solution to plutonium oxide would be accomplished in the same manner identified in the ORNL preconceptual design study for plutonium-238 production support operations (Wham et al. 1998). Plutonium oxide conversion would occur in Cell 144. The plutonium nitrate solution would be treated to adjust the plutonium oxidation state and then precipitated as an oxalate. The plutonium oxalate would be washed with dilute acid to remove entrained liquor, transferred to a boat and calcined at the required temperature. The required oxygen-16 exchange would be done as part of the calcination step. The oxide would be packaged in a manner to assure maintenance of the desired degree of oxygen-16 exchange. The packaged material would undergo nondestructive assay analysis for accountability control and then be placed into storage pending packaging for shipment.

A.3.5 Plutonium-238 Storage Description

Plutonium-238 oxide would be stored in racks in Cell 143 on the -35-foot level. Storage quantities would be expected to be small, since shipments would be made to LANL on a regular basis. Alternatively, the special nuclear material storage vault located in Rooms 428 and 429 on the 21-foot level could be used. In addition, any of several hardened rooms within FMEF could be modified to be vault-type rooms for storage of plutonium-238 oxide packaged in shipping containers awaiting shipment.

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